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Quantum Oscillation of the Tunneling Conductance in Fully Epitaxial Double Barrier Magnetic Tunnel Junctions

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We investigated spin-dependent tunneling conductance properties in fully epitaxial double MgO barrier magnetic tunnel junctions with layered nanoscale Fe islands as a middle layer. Clear oscillations of the tunneling conductance were observed as a function of the bias voltage. The oscillation, which depends on the middle layer thickness and the magnetization configuration, is interpreted by the modulation of tunneling conductance due to the spin-polarized quantum well states created in the middle Fe layer. This first observation of the quantum size effect in the fully epitaxial double barrier magnetic tunnel junction indicates great potential for the development of the spin-dependent resonant tunneling effect in coherent tunneling regime.

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Spintronics, which introduces spins of electrons as an additional degree of freedom in electronics, has been creating widespread interest due to its large potential implication in the creation of electronic devices based on a new principle of operation. One of the most successful discoveries in the research field of spintronics is the large tunnel magnetoresistance (TMR) effect at room temperature (RT) in a magnetic tunnel junction (MTJ), consisting of $\mathrm{FM}_1/I/\mathrm{FM}_2$ structure where FM_1 and FM_2 are ferromagnetic layers and I is an ultrathin insulator [1,2]. The MTJ has been receiving increasing attention due to its possible technological applications, such as a memory cell in magnetic random access memories (MRAMs).

The effective utilization of coherent tunneling effect for the large enhancement of TMR ratio ($\sim$1000%) in a fully epitaxial Fe(001)/MgO(001)/Fe(001) system has been suggested by numerical calculations [3,4]. The origins of this large TMR enhancement are attributed to the symmetry of Bloch states of Fe(001) electrode and relevant evanescent states of MgO(001) barrier. In the case of Fe[001] direction, only Δ_1 band (s character) electrons in the majority spin channel couple efficiently with decaying sp states in the MgO(001) barrier and contribute to the large tunneling conductance in the parallel magnetization configuration. On the other hand, the absence of Δ_1 band states and the rapid decay of other symmetric bands, e.g., $\Delta_5(pd)$, $\Delta_2(d)$, and $\Delta_{2'}(d)$, in the MgO barrier for the minority spin channel lead to the small conductance in the antiparallel magnetization configuration. Recently, Yuasa *et al.* [5] and Parkin *et al.* [6] achieved considerably large TMR ratios which largely surpass the maximum TMR value observed in the MTJs with an AlO_x barrier [7]. This discovery should be a breakthrough for the realization of gigabit-scale high-density MRAMs.

On the other hand, spin-dependent resonant tunneling effect in double barrier MTJs (DMTJs) consisting of $\mathrm{FM}/I/\mathrm{FM}_2/I/\mathrm{FM}_3$, where spin-dependent quantum well (QW) states are created in the ultrathin FM_2 layer, is one of fascinating spin-dependent phenomena. Zhang *et al.* [8] and Kishi *et al.* [9] investigated theoretically the conductance and TMR properties subjected to the electric field for the DMTJs with an ultrathin middle ferromagnetic layer and pointed out that the tunneling conductance oscillates and exhibits peaks at resonant voltages, leading to the enhancement of TMR effect. The spin-polarized QW effect through an ultrathin nonmagnetic [10,11] or magnetic [12] layer using the band structure of metals have already been reported. However, since the reflection of electrons at the interface of metal junctions is imperfect, a double barrier structure is indispensable for the complete confinement of electrons and the realization of the resonant tunneling effect. In this work, we first demonstrate the quantum oscillation of the tunneling conductance observed in fully epitaxial DMTJs.

Pseudo-spin-valve type DMTJs, consisting of MgO seed layer (10)/Fe(50)/ MgO(2)/Fe(t)/MgO(2)/Fe(15) (the numbers are film thickness in nm), were deposited on single crystal MgO(001) substrates using molecular beam epitaxy with a base pressure of 5×10^{-8} Pa. The designed thickness of the middle Fe layer, monitoring by a quartz oscillator, was varied from 1.0 to 1.5 nm. The details of the sample preparation are shown in Ref. [13]. Epitaxial growth of each layer was confirmed by observing a reflection high-energy electron diffraction (RHEED) pattern during deposition and after annealing treatment (Fig. 1, $t = 1.5$ nm). All layers except for the middle Fe layer exhibit narrow streaks indicating a two-dimensional epitaxial growth mode. On the other hand, diffusive spots indicating a three-dimensional growth mode are observed for the as-deposited middle Fe layer at RT as previously reported [14]. However, after annealing treatment at 200 °C, the diffusive spots change to tiny spots and narrow streaks appear [see Fig. 1(d)].

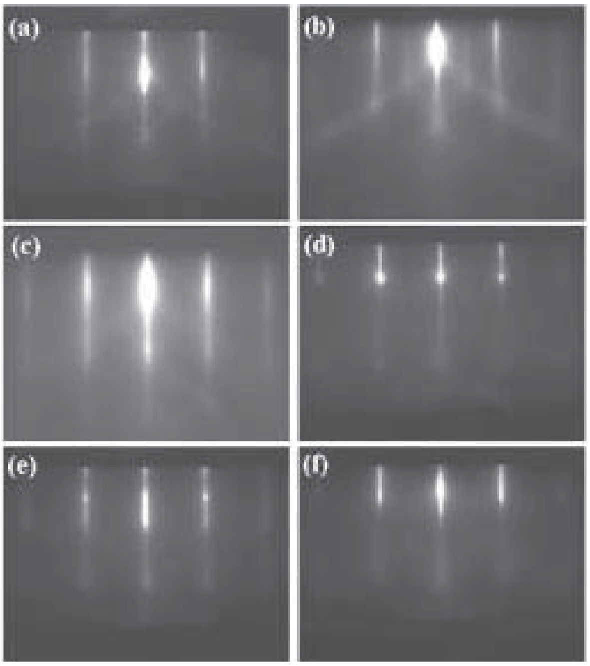

FIG. 1. RHEED patterns of the (a) MgO seed layer, (b) bottom Fe electrode, (c) first MgO barrier, (d) middle Fe layer, (e) second MgO barrier and (f) top Fe electrode in the double barrier MTJ. The electron beam is incident along the [100] and [110] azimuth in the MgO and Fe layer, respectively.

For clarifying the condition of the middle Fe layer, cross-sectional transmission electron microscopy (TEM) analysis was carried out. Figure 2 shows the TEM image for the DMTJ with $t = 1.5$ nm. From this TEM image and the low magnification image (not shown here), we can see that the middle Fe layer forms layered islands (dashed line area) of 20–60 nm in diameter and 5.3 nm in height, which is 3.3 times thicker than the designed thickness. The sharp streaks and the tiny spots observed in Fig. 1(d) come from the surface of the MgO barrier layer and the Fe islands, respectively. It is noteworthy that the isolated Fe islands are completely covered with the second MgO barrier and the interfaces of MgO/Fe islands/ MgO are very flat. Since the tunneling currents pass through the Fe islands selectively due to the thick barrier thickness (~4 nm) beside the islands, the whole junction can be considered as parallel-connected DMTJs.

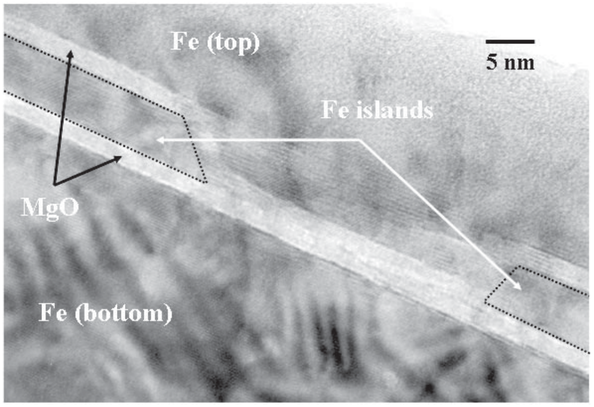

FIG. 2. Cross-sectional TEM image of the double barrier MTJ with the designed middle layer thickness of $t = 1.5$ nm.

For the measurement of transport properties, samples were patterned into junctions of 10×10 μm^2 in size by a successive micro-fabrication process with the combination of both electron-beam lithography and Ar ion milling. Both TMR and current-voltage (I-V) curves were measured using a dc four-probe method. We define the TMR ratio as $(R_{\rm AP} - R_{\rm P})/R_{\rm P}$, where $R_{\rm AP}$ and $R_{\rm A}$ are the resistance of the antiparallel and parallel magnetization configurations between the top and bottom thick Fe electrodes, respectively. During the measurement, tunneling electrons flow from the bottom to the top electrode with $V > 0$.

Figure 3 shows the representative TMR curve of the DMTJ with $t = 1.5$ nm measured at 50 K. A magnetic field (H) was applied along the Fe[110] ($\parallel$ MgO[100]) direction (The TMR ratio measured in the easy axis direction was smaller because of the similar coercivity of Fe electrodes). In spite of the imperfect antiparallel magnetization state due to the hard axis measurement direction and random magnetization configuration of Fe islands in the small magnetic field region (we confirmed it from the TMR measurement in the Fe(50)/MgO(2)/Fe(1.5)/MgO(2)/Cr(10) structure), we observe the large TMR ratio of 136% with the resistance area product (RA) of 18 kΩ μm^2 which is more than 5 times higher than that for the single barrier MTJ with the same MgO barrier thickness (not shown here). The origin of the RA enlargement is caused by the reduction of the actual junction size because of the formation of Fe islands.

Figure 4(a) demonstrates the conductance (dI/dV) curves measured at 4.5 K for the DMTJs with various Fe

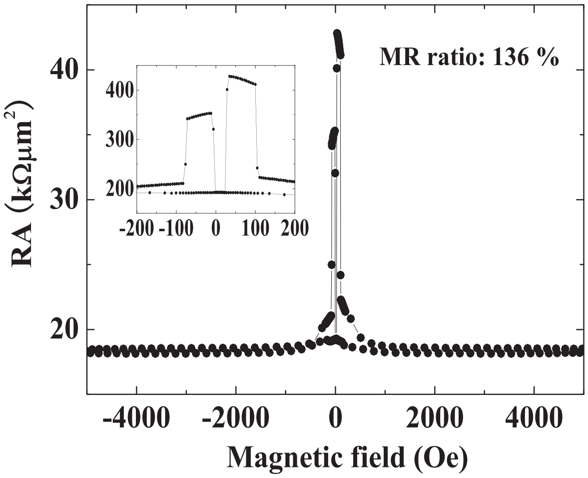

FIG. 3. Representative TMR curve of the DMTJ for the same sample shown in Figs. 1 and 2. The inset shows the same curve in a small magnetic field range.

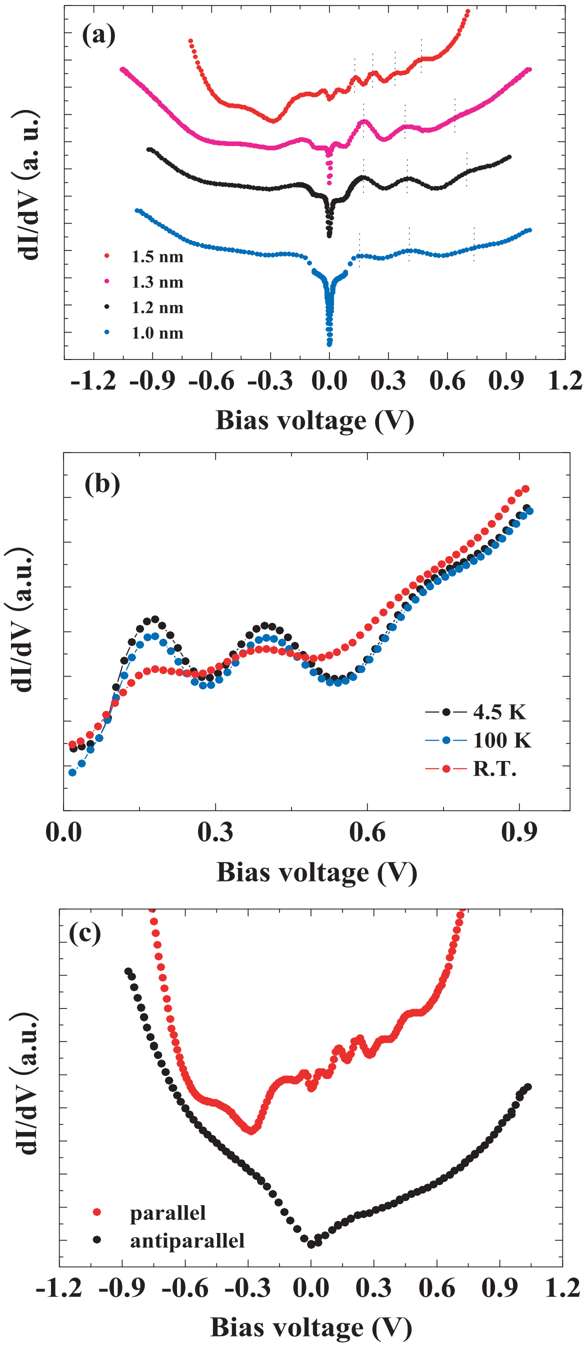

FIG. 4 (color). (a) Designed middle layer thickness (t) dependence of conductance curves in the parallel magnetization configuration (measured at 4.5 K). (b) Temperature dependence of conductance curves for $t = 1.2$ nm in the positive bias direction. (c) Magnetization configuration dependence of conductance curves for $t = 1.5$ nm (measured at 4.5 K). The parallel and antiparallel magnetization configurations were realized by the applying magnetic field of 5000 and 50 Oe, respectively.

middle layer thicknesses (designed thickness, $t =$ 1.0–1.5 nm). We clearly observe the oscillations of the conductance in the positive bias direction. We must pay some attention to evaluating the conductance curve of the MTJ with a MgO barrier. First, the influences of magnon and phonon assisted tunneling could be observed in the low bias region under 0.2 V [12]. Second, the local minimum arises at $V = 0.3$ V in the MTJ with a MgO barrier [13], although the origin of this dip is not clear at this stage. From these perspectives, it is difficult to assess the first peak at around $V = 0.2$ V as a part of the oscillation, however, such a sharp peak has not been observed in a single MgO barrier junction [13]. Furthermore, we clearly observe the third and fourth peaks at $V > 0.3$ V for the sample with $t = 1.5$ nm. In addition, since the bias voltage where the conductance shows the maximum changes to a higher value with decreasing the middle layer thickness, the origin of these oscillations is attributed to the effect of the quantum well states created in the layered Fe islands. Although similar oscillations of the conductance have been reported by Nagahama *et al.* in Cr(001)/ultrathin Fe(001)/AlO_x/CoFe structure [12], the oscillatory features in their results were very small in the first derivative of I-V curves. They attributed this weak amplitude of the oscillation to the short mean free path and spin diffusion length in the ferromagnetic metal. However, we can clearly identify the oscillatory feature even in the first derivative of I-V curve. This enhancement is assumed to be due to the strong confinement effect by the MgO barriers. However, even in our structure, the oscillations of resistance and TMR ratio as a function of the bias voltage are still very slight. Further reduction of the middle layer thickness and the prevention of the island formation should be required for larger enhancement.

Figure 4(b) shows the temperature dependence of the conductance curves of the DMTJ with $t = 1.2$ nm (actual thickness was estimated to be 2 nm from the cross-sectional TEM analysis) in the positive bias direction. Although the amplitude of the oscillation attenuates with increasing temperature, we can observe the oscillatory feature even at RT. On the other hand, for the case of $t = 1.5$ nm, the oscillatory feature disappears at RT due to its small energy gap between the quantum levels.

Figure 4(c) is the magnetization configuration dependence of the conductance curves of the DMTJ with $t =$ 1.5 nm. The oscillatory feature is observed only in the parallel magnetization configuration. This result indicates that the QW states are created by the only one of the two spin bands. From the simple consideration of the coherent tunneling mechanism in Fe(001)/ MgO(001)/Fe(001) structure, we can speculate that these spin-polarized QW states are ascribed to the majority Δ_1 band state in the Fe(001) electrode. The majority $\Delta_{2'}(d)$ and $\Delta_5(pd)$ bands also have prospects of QW formation, however, since the top of these bands resides at 0.14 eV above the Fermi energy, the oscillatory peaks observed at high voltage region more than 0.2 V cannot be explained. For this reason, the oscillation should be observed only in the parallel magnetization configuration.

The reason the oscillations are observed only in the positive bias direction is speculated to be the asymmetry of the interfacial structure. H. L. Meyerheim *et al.* pointed out that the existence of an FeO polluted layer, which can be formed when a MgO is evaporated on an Fe electrode, significantly affects to the spin-dependent tunneling effect [15]. When a negative bias voltage is applied, the tunneling conductance reflects the unoccupied electronic states of the bottom Fe electrode through the middle Fe layer. If the FeO layer exists on only the top side of the bottom and middle Fe layers adjacent to the MgO barrier, not only Δ_1 majority spin band but also other symmetric bands can contribute to the tunneling conductance for the negative bias voltage application, making the QW effect obscure. On the other hand, in the positive bias direction, the clean Fe/ MgO interfaces work as a filter for electrons with different symmetric bands. This selective coherent tunneling of the majority Δ_1 band electrons conserves the QW effect only in the positive bias direction. The strong asymmetric bias voltage dependence of the TMR ratio observed in the DMTJs [14] is also attributed to the same reason. The zero-bias dip observed in the conductance curves, which becomes larger with decreasing the thickness, may be the influence of Coulomb blockade effect resulting from the reduction of the island's size.

In the last part of this Letter, we evaluate the prospective thickness of the middle Fe layer from the observed resonant peaks and the band structure of Fe(001) with a little arithmetic. In general, the quantization condition is expressed as

$$2kt + \Phi_B + \Phi_T = 2\pi n \qquad (n = \text{integer}), \tag{1}$$

where k describes the wave vector of the majority spin Δ_1 band of Fe(001) and Φ_B and Φ_T are the phase changes of the electron wave function upon reflection at the bottom-side and top-side Fe/MgO interfaces. For simplicity, we neglect these phase changes here. For the case of $t = 1.5$ nm, we observed 4 resonant peaks at the bias voltage of $V = 0.13$, 0.22, 0.33 and 0.47 V. From the band dispersion of the majority Δ_1 band of Fe(001) [16], the wave number (Δk) change in this energy range is 0.08 $[2\pi/a]$ (a is the lattice constant of Fe: 0.287 nm). If we assume that the energy interval of quantum levels is approximately the same in the observed energy range, we can evaluate the prospective thickness $t = 4\pi/\Delta k = 4a/0.16 = 7.2$ nm. This is thicker than the actual thickness of the Fe islands observed in Fig. 2 by 2 nm. This discrepancy may be caused by the neglect of the condition of Fe/MgO interfaces. Furthermore, as mentioned above, since the MTJ with a MgO barrier shows a characteristic conductance curve with a dip at $V = 0.3$ V [13], we need to subtract these backgrounds for the accurate estimation of the oscillation periods. When we neglect the dip at $V = 0.3$ V in the conductance curve of $t = 1.5$ nm, the prospective thickness is reevaluated as $t = 3\pi/\Delta k = 5.2$ nm, which is in good agreement with the TEM observation.

In summary, we investigated the transport properties of the fully epitaxial MgO double barrier magnetic tunnel junctions with layered Fe islands as a middle layer. Clear oscillations of the tunneling conductance were observed for various middle layer thicknesses only in the parallel magnetization configuration. These oscillations reflect the modulation of the tunneling conductance by the spin-polarized QW states created in the middle Fe islands. This is the first observation of the quantum size effect in the fully epitaxial double barrier magnetic tunnel junctions. Further improvement of the quality of the quantum well layer will provide the prominent QW effects and the realization of the spin-dependent resonant tunneling effect.

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